

COKE FORMATION AND CHARACTERIZATION DURING THERMAL TREATMENT AND HYDROCRACKING OF LIAOHE VACUUM RESIDUE

Zong-xian Wang, Ai-Jun Guo, Guo-he Que

National Heavy Oil Processing Laboratory, University of Petroleum
Dongying, Shandong 257062, P. R. China

The physical and chemical properties and structural compositions of Liaohe residuum were studied. The coke formation in the processes of thermal conversion and slurry catalytic hydrocracking reaction of the residuum was subsequently investigated. Then, the relationship between coke formation tendency and physicochemical compositions of the residuum was studied. The effect of initial coke formation on the propagation of coke formation was also studied. Using high-precision microscope and FTIR, the formed coke was characterized. With increasing processing severity, the coke formed in the bulk of reaction fluid system is changing from fine particulate form to coke cluster form. Initially formed coke seems to promote coke formation and growth of coke clusters during thermal treatment under low pressure. The onset of coke formation is quite closely related to the ability of reaction system to peptize coke precursors.

KEY WORDS: coke formation, thermal treatment, hydrocracking, vacuum residue.

INTRODUCTION

Coke formation has long been a concern of petroleum refiners and researchers because petroleum residue processing is often limited by coke formation. This limitation is very severe in the cases of visbreaking, catalytic hydrotreating and catalytic hydrocracking, in which no appreciable amount of coke can be tolerated because of the need to flow freely through coils or catalyst-bed and the need to keep catalysts active. In the FCC or RFCC and slurry-bed catalytic hydrocracking (SBCH) processes, coke formation may deactivate catalysts and decrease the depth of upgrading. Even the delayed-coking process would be limited by coke formation in heating coils. Hence, the coke formation is one of key factors influencing petroleum residue upgrading.

In addition to processing conditions, the physical and chemical properties and structural composition of petroleum residues dictate the action of coke formation (1-4). A vacuum residue(VR) with low H/C atomic ratio and high carbon residue may be of high propensity to produce large amount of coke, to which much attention has been paid by RFCC, Coking and SBCH processes. The inducing period of coke formation is not only related to carbon residue value but also to the miscibility of residue sub-fractions and particularly to the ability for vacuum residue system to peptize its asphaltene and coke precursors (5-7). Evidently, the long inducing period of coke formation is relevant to visbreaking and hydrotreating processes. Therefore, the study on coke formation is of significance in searching for coke inhibition procedures.

This work is intended to take Liaohe vacuum residue as a specimen to study, in detail, its physical and chemical properties and structural composition, and to investigate coke formation during its thermal, hydrothermal conversion and slurry-bed catalytic hydrocracking.

EXPERIMENTAL

Sample

Liaohe vacuum residue was collected from Liaohe Petrochemical Plant in March 1996; Gudao VR was collected from Shengli Oil Refinery. Their general properties are listed in Table 1.

Analytical Procedures

Isolation of Six Sub-fractions

Liaohe and Gudao vacuum residues were chromatographically separated into six fractions, i.e. light oil (F1), heavy aromatic fraction (F2), light resin (F3), middle resin (F4), heavy resin (F5) and n-pentane asphaltene (F6) by using a procedure described in reference (8)

Ultimate Analysis and Molecular Weight Measurement

Carlo Erba 1160 elemental analyzer was used for C, H, N analysis; atomic absorption method was used to determine Ni, V, Fe and Ca contents. Average molecular weight was measured by using VPO method (benzene as solvent, 45 °C) with Knauer molecular weight analyzer.

FTIR and Microscopic Characterization of Petroleum Coke

FTIR analysis of coke was conducted at Nicolet Magna 750 IR analyzer, a mold flat of coke and KBr mixture as sampling. An OLYMPAS HS-2 microscope was used to visualize the coke formed in the bulk of reaction system.

Liaohe VR Thermal and Hydrothermal Conversion and Catalytic Hydrocracking

The experiments were carried in a 100ml FDW-01 autoclave reactor with an up-and-down stirrer at 120 times of reciprocation per minute. Initial pressure was 5.0Mpa N₂ for thermal conversion and 7.0MPa H₂ for hydrothermal conversion and catalytic hydrocracking. Catalyst used in hydrocracking reaction was Mo based oil soluble additive (ca.200 PPM Mo in reaction feed), and was pre-sulfided by elemental sulfur at 320°C for 30minute after mixing with Liaohe VR (S/Mo atomic ratio=3/1). Reaction temperature (430°C) was reached within 25minute from room temperature. After 1 hr. reaction time, the reactor was quenched (cooled) to room temperature, the reactor gas was vented, and toluene slurry was prepared from the reactor contents. Any solids adhering to the reactor walls or internals was carefully scraped off. The slurry was centrifuged into the toluene insoluble (TI or coke) and the toluene soluble; the toluene insoluble was then washed (extracted) with boiling toluene and separated by using quantitative filter paper, and then dried and weighed. The toluene soluble was distilled into several fractions. The data are listed in table 4.

RESULTS AND DISCUSSION

Relation Between Feed Properties and Coke Formation

The data in table I show that Liaohe VR, in contrast to Gudao VR, is high in viscosity, CCR, heavy metals, nitrogen content, aromaticity (f_A) and the ratio of aromatic ring number to naphthenic ring number (R_A/R_N). This indicates that Liaohe VR will generates more coke than Gudao VR under high processing severity (e.g. delayed coking and RFCC).

However, the SARA analysis (group composition) is similar for these two VR, for example, the oil fractions (saturates+aromatics) are 47.7% and 49.3% for Liaohe VR and Gudao VR respectively. Even the n-heptane asphaltene (nC7-At) content of Liaohe VR is less than that of Gudao VR. Even the oil fraction of Liaohe VR has lower density, viscosity, pour point, CCR, sulfur, nitrogen and metal contents, contrast to the oil fraction of Gudao VR.

Therefore, it can be inferred that the qualities of resin and asphaltene fractions of Liaohe VR must be much inferior to those of Gudao VR. It is verified by further analysis of six sub-fractions of these two VR, as seen in table 3. The nitrogen, nickel and CCR in heavy resin and asphaltene account for 69.1%, 78.6% and 74.1% respectively of total N, Ni and CCR in Liaohe VR, while the corresponding values are only 61.0%, 61.0% and 63.0% for Gudao VR. The aromaticity and R_A/R_N ratio of Liaohe resin and asphaltene are much higher than those of Gudao resin and asphaltene, indicating that the cracking performance of Liaohe resin and asphaltene must be quite poor.

Besides, there exists a great difference in molecular weight distribution in Liaohe VR six sub-fractions. The ratio of asphaltene molecular weight to oil molecular weight for Liaohe VR is about 9.5, while that for Gudao is only about 4.6. The gap between properties of oil fraction and resin-asphaltene fraction of Liaohe VR is much wider than that of Gudao VR, which implies that the system of Liaohe VR is not harmonious, and its colloidal stability must be very low. All those above show that Liaohe VR is kind of inferior heavy oil; the propensity to coke formation is much higher than Gudao VR during processing. A preliminary thermal treatment test has shown that the inducing period of coke formation of Liaohe VR is shorter than that of Gudao VR.

Distribution of Products in Thermal Treatment and Hydrocracking of Liaohe VR

Table 4 lists 450°C⁺ fraction, 450°C⁺ cracked residue, coke content in reaction products and

pentane asphaltene content in cracked residue. With reaction temperature being increased, 450°C fraction yields increases for all three series of reactions, and the increment is the highest for thermal reaction and the least for catalytic hydrocracking with Mo based catalyst. But the conversion to light products is the highest for the latter process under unit coke formation condition. Under the reaction condition of 430°C and reaction time 1 hr, the coke formation is less than 1% and conversion reaches up to 50% for the latter process; while coke formation is much greater than 1% as conversion reaches 50% for other two series of reactions of Liaohe VR. The coke formation increased rapidly with temperature for thermal treatment of Liaohe VR, but quite slowly for catalytic hydrocracking. The asphaltene contents in three reaction systems are changed in the similar tendency with cracked residue proportions. The molecular weight of asphaltene decreases with increasing reaction temperature for all these three reaction systems. The decrease of asphaltene molecular weight for thermal treatment is most eminent.

Characterization of Coke Formation by Microscopic Analysis

Reacted materials were sampled from the bulk of reaction system with thin glass plate as the samplers and then visualized by using an OLYMPAS HS-2 microscope. Three series of microphotographs ($10\times40\times3.3$) were taken of reacted materials from three series of reaction systems of Liaohe VR thermal treatment, hydro-thermal treatment and catalytic hydrocracking. The microscopic analysis showed that there was yet little coke at 420°C for hydrocracking system. With temperature increasing, the coke formed in the bulk of reaction system was changing from fine dispersed particle forms to small clusters(about 0.1-0.2% coke contents), corresponding temperatures being at 425, 415 and 400°C for thermal treatment, hydro-thermal treatment and catalytic hydrocracking respectively; and further to coke clusters corresponding temperatures being at 430, 420, 415°C respectively.

Characterization of Coke by FTIR

Fourier Transition Infrared spectrometry (FTIR) is a useful technique for characterizing organic functional groups of organic substances. Naturally it can be used to characterize the coke formed in reaction system and to describe the changes of different organic function groups, such as aromatic carbon, paraffinic carbon and substitution extent of aromatic ring periphery, of coke with increasing reaction severity.

Aromatic C=C double bond vibrates at 1600 cm^{-1} aromatic C-H bond vibrates at 3030 cm^{-1} , and 750 cm^{-1} , 810 cm^{-1} , 870 cm^{-1} . Paraffinic C-H vibrates at 2920 cm^{-1} , 2860 cm^{-1} , 1460 cm^{-1} and 1380 cm^{-1} . Here, the relative vibrating strength (A_x), i.e., the ratio of vibrating strength (A_x) of function groups to C=C vibrating strength (A_{1600}), was used to describe the relative changes of coke function groups and to reveal the way in which chemical structures change. The relative vibrating strengths of several organic function groups of coke formed at different severity in Liaohe VR reaction systems were tabulated in table 5. With temperature increasing, A_{2920} and A_{2860} seemed gradually decreasing, showing the paraffinic portion of coke decreasing and aromaticity relatively increasing. A_{870} , A_{810} and A_{750} increased with temperature increasing, showing that constitution extent of aromatic ring was decreasing and side chain cracking occurred. Therefore, FTIR is an effective means for characterization of coke formation during VR processing.

Mechanism of Initial Coke Formation and Its Effect

Vacuum residue is a colloidal system with asphaltene and heavy resin as dispersed phase and oil fraction and light resin as media. If physical and chemical properties of dispersed phase were much different from those of media constituents, the media would not properly peptize the asphaltene, and the colloidal system would not be stable. For example, asphaltene can be precipitated by n-pentane because pentane dilutes the colloidal system and makes the media less aromaticity, low average molecular weight and low polarity. When heating disturbs vacuum residue system, the molecules move fast, the micelle made up of asphaltene and media would become loser, some of asphaltene molecules may bump off the micelle cages to coalesce. The vacuum residue colloidal system can also be disturbed by some fine solid polar particles due to the adsorption of asphaltene molecules to these particles. At high heating severity, some weak bonds in asphaltene molecules may crack. Asphaltene micelles may break apart; some asphaltene molecules may physically coalesce or chemically interact to form large molecules. At the same time, the ability for media to peptize asphaltene molecules becomes lower due to light fractions

increasing, thus causing asphaltene molecules to coalesce and to be precipitated.

All these factors can account for the initial coke formation of vacuum residue reaction system. In fact, the initial coke is the physical-chemically-coalescent phase of asphaltene molecules, whose properties are very similar to those of asphaltene---coke precursor. Hence initial coke may have high an affinity for asphaltene molecules and promote coalescence of asphaltene molecules and coke formation.

In order to testify the effect of the initial coke on reaction system, Liaohe VR with some pre-added coke powders was subjected to thermal treatment and hydrocracking tests. The thermal treatment under atmospheric nitrogen gas was conducted in a quartz tube reactor (30 ml) heated by an electric tin bath, other tests were carried out in an autoclave of 100 ml under 7MPa hydrogen gas. The results were tabulated in table 6.

Coke formation was about 0.73% for Liaohe VR thermal treatment under conditions of 406°C 2hr and nitrogen atmosphere. When 0.5% coke was pre-added in the thermal treatment system, total coke was about 1.93%, net coke formation was 1.20%, net coke increment was about 0.47% contrast to blank test without pre-added coke; 1% pre-added coke promoted 1.22% net coke formation compared with the blank thermal treatment. It was obvious that initial coke could considerably promote coke formation during thermal treatment of vacuum residue under low pressure(a relatively open system). In hydrothermal treatment of VR, it seemed, superficially, that pre-added coke was of little influence on coke formation. In fact, the Ni, Fe in pre-added coke might act as hydrocracking catalysts, the catalytically active hydrogen could saturate the coke precursor free radicals and chemically inhibit condensation of these free radicals, thus inhibit the coke formation. The counteraction between the coke promotion by initial coke and the coke inhibition by heavy metals in the coke makes total coke formation similar to the coke formation in virgin hydrothermal treatment. In the catalytic hydrocracking process, pre-added coke could markedly inhibit net coke formation. In addition to activating hydrogen, the pre-added coke and Mo sulfide solid particles were of high affinity for asphaltene free radicals, and made these coke precursor radicals adsorbed around them. Thus, the active hydrogen on catalysts could instantly terminate coke precursor radicals; thus the coke inhibition efficiency was enhanced. The performance for catalyst particles to enrich asphaltene-coke precursors around them and to promote the utilization of active hydrogen may be the important factors for slurry-bed catalytic hydrocracking of heavy oils to effectively inhibit coke formation.

CONCLUSION

The SARA group composition of Liaohe VR is similar to that of Gudao VR, the nature of its oil fraction is even better than that of the Gudao VR counterpart, but its resin and asphaltene fractions are much inferior to those of Gudao VR. The uniformity of Liaohe VR system is lower than that of Gudao VR, its colloidal system is not stable, and the coke formation tendency is high during further processing.

With processing severity increasing, coke formation in VR reaction system is developed from dispersed fine particles to coke clusters. FTIR technique can effectively characterize the evolution of aromatic part and paraffinic part of the coke formed at different severity.

The initial coke could promote coke formation during low-pressure thermal treatment of VR. During hydrothermal and hydrocracking treatments, pre-added coke seems to promote catalysis.

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Table 1 Properties of Liaohe vacuum residue (VR)

Property	Liaohe VR	Gudao VR
Density(20°C), g./cm ³	0.9976	0.9998
Viscosity(100°C)/mm ² .s ⁻¹	3375	1710
Pour point, °C	42	41
Flash point, °C	312	327
Carbon residue, %	19.0	15.6
Elemental composition		
C, %	87.0	85.4
H, %	11.4	11.4
S, %	0.43	2.52
N, %	1.08	0.80
H/C(Atomic ratio)	1.50	1.60
Total Metal/PPM	258.6	131.6
Ni, PPM	122.6	48.0
V, PPM	2.9	2.2
Fe, PPM	37.5	13.8
Ca, PPM	95.6	33.8
Ash, %	0.056	0.026
SARA fractions:		
Saturates, %	17.1	14.5
Aromatics, %	30.3	34.8
Resins, %	50.2	47.2
nC7-Asphaltene, /%	2.1	3.5
Structural Parameters		
F _A	0.267	0.181
F _N	0.258	0.330
R _A /R _N	0.93	0.47

Table 2 Properties of oil fraction of Liaohe VR

Properties	Oil fraction of Liaohe VR	oil Fraction of Gudao VR
Density(20°C), g/cm ³	0.9392	0.9558
Viscosity(100°C), mm ² .s ⁻¹	84.1	103.0
Pour point, °C	36.0	39.0
Carbon residue, %	3.5	5.8
MW	630	860
Elemental composition		
C, %	87.2	85.0
H, %	12.3	11.9
S, %	0.34	2.0
N, %	0.33	0.45
H/C(Atomic ratio)	1.68	1.67
Metals		
Ni, PPM	1.3	6.0
V, PPM	2.4	/
Ca, PPM	1.1	19.8

Table 3 Properties and composition of six fractions of Liaohe VR

Property	F1	F2	F3	F4	F5	nC ₅ -At
<i>Liaohe VR:</i>						
Yield, %	36.1	11.6	16.8	9.0	12.3	14.2
MW	590	810	1050	1380	2240	5970
f _A	/	/	0.329	0.357	0.371	0.509
f _N	/	/	0.201	0.153	0.151	0.082
R _A /R _N	/	/	1.5	2.2	2.4	6.2
H/C	1.75	1.48	1.44	1.41	1.38	1.19
N, %	0.16	0.85	1.42	1.70	1.78	1.96
N/N _T , %	2.1	10.8	18.0	21.6	22.6	24.9
Ni, ppm		1.3	197.3		225.2	502.3
Ni _T /Ni _T , %		0.2	21.3		24.3	54.3
CCR, %		3.5	23.2		31.1	45.3
CCR _T /CCR _T , %		3.4	22.5		30.2	43.9
<i>Gudao VR:</i>						
Yield, %	34.9	14.2	15.6	7.9	8.8	15.9
MW	800	1000	1370	1760	2430	3920
f _A	/	/	0.285	0.290	0.297	0.437
f _N	/	/	0.149	0.182	0.166	0.128
R _A /R _N	/	/	1.8	1.5	1.7	3.3
H/C	1.83	1.50	1.51	1.49	1.48	1.29
N, %	0.10	0.75	1.17	1.37	1.43	1.48
N/N _T , %	4.2	12.9	21.9	13.0	19.7	28.3
Ni, ppm	0.6	9.7	109.1	43.7	49.6	127.3
Ni _T /Ni _T , %	0.4	2.9	35.7	7.2	12.0	41.8
CCR, %	2.4	15.9	21.7	23.6	21.7	41.8
CCR _T /CCR _T , %	4.8	12.9	19.3	10.6	14.5	37.9

MW - molecular weight t. N/N_T - the ratio of nitrogen in fraction to total nitrogen in VRNi_T/Ni_T - the ratio of nickel in fraction to total nickel in VRCCR/CCR_T - the ratio of carbon residue of fraction to total carbon residue in VRTable 4 Yields of conversion products under three series of reaction conditions
(Reaction time 1hr)

Reaction condition		<450°C product, %	>450°C cracked residue, %	Coke, %	nC ₅ -At in cracked residue	
					%	MW
N ₂ 5MPa	400	37.7	62.2	0.1	/	/
	410	44.4	54.2	1.4	24.6	3349
	420	67.1	32.9	10.9	15.8	2829
	430	62.8	17.7	19.5	9.2	1499
H ₂ 7MPa	400	28.27	71.68	0.05	/	/
	410	40.90	59.00	0.10	23.1	3837
	420	50.20	46.60	3.20	17.9	2989
	430	61.40	30.78	7.92	13.3	1979
H ₂ 7MPa Mo 200ppm	400	18.95	81.00	0.05	/	/
	410	25.60	74.31	0.09	/	/
	420	35.54	64.24	0.22	15.8	3477
	430	50.19	49.13	0.68	14.8	2932
	440	53.79	42.0	4.21	12.2	2063

Table 5 Relations between coke FTIR Data and reaction conditions

Coke samples	A ₂₉₂₀	A ₂₈₆₀	A ₃₀₃₀	A ₈₇₀	A ₈₁₀	A ₇₅₀
N2						
410°C	0.780	0.493	0.060	0.271	0.206	0.185
420°C	0.650	0.426	0.095	0.283	0.243	0.241
430°C	0.273	0.210	0.098	0.349	0.312	0.294
H ₂ , Mo200ppm						
425°C	0.574	0.385	0.068	0.280	0.152	0.149
430°C	0.375	0.256	0.058	0.278	0.210	0.184
435°C	0.265	0.176	0.060	0.214	0.215	0.196
440°C	0.183	0.155	0.030	0.390	0.422	0.406

Table 6 Effect of initial coke on coke formation of reaction system

Reaction condition	sample	<450°C product, %	>450°C cracked residue, %	** coke, %	*** coke increment, %
* N ₂ (blowing), 2hr					
406°	LHVR	/	/	0.73	
406°	LHVR+0.5%coke	/	/	1.20	0.47
406°	LHVR+1.0%coke	/	/	1.95	1.22
H ₂ , 7MPa, 2hr					
412°	LHVR	53.37	43.70	2.93	
412°	LHVR+0.5%coke	54.06	42.50	2.94	0.01
H ₂ , 7Mpa, 2hr Mo, 200ppm					
425°	LHVR	56.87	41.50	1.63	
425°	LHVR+0.5%coke	52.96	45.40	1.14	-0.49

* conducted by using a quartz tube reactor with N₂ as a blowing gas;

** net coke formation, i.e., total coke minus added coke.

*** coke increment on the base of blank test.

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